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CLAIMS

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[Claim(s)]

[Claim 1] (1) The ionomer layer which consists of an ionomer, In a list, according to the (2) and (A) transition metal catalyst The crystalline ethylene and alpha olefin copolymer, and the (b) consistency whose (a) consistency by which the polymerization was carried out is 0.89 - 0.94 g/cm<sup>3</sup> are less than [ 0.89g //cm ] three, and crystallinity consists of 40% or less of amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity. The crystalline ethylene and the alpha olefin copolymer the 1st resin constituent whose consistencies are less than three 0.930 g/cm, and whose (c) consistency the polymerization was carried out [ the consistency ] by the (B) transition metal catalyst are 0.92 g/cm<sup>3</sup> at least, (d) Ethylene-vinylacetate copolymer, From the ethylene copolymer chosen from the group which consists of an ethylene acrylic-acid copolymer and an ethylene ethyl acrylate copolymer The 2nd resin constituent which contains (c) crystallinity ethylene and 95 - 50 % of the weight of alpha olefin copolymers, and 5 - 50 % of the weight of (d) ethylene copolymers based on the sum total weight of crystalline ethylene and the alpha olefin copolymer (above-mentioned [ c ]), and the ethylene copolymer (above-mentioned [ d ]) And the resin layered product characterized by the resin constituent layer which consists of a resin constituent chosen from the group which consists of the 3rd resin constituent which comes to carry out graft denaturation of the 1st resin constituent of (C) above or the above-mentioned 2nd resin constituent with unsaturated carboxylic acid adjoining and existing.

[Claim 2] (1) The resin layered product characterized by a \*\*\*\* resin layer adjoining and existing in such sequence if it chooses from the group which consists of the ionomer layer which consists of an ionomer, the resin constituent layer which consists of the (2) 3rd resin constituent, (3) nylon, and an ethylene-vinylalcohol copolymer.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to a resin layered product. It is related with the resin layered product which consists of an ionomer layer, and this and an adhesive good resin constituent layer in more detail.

[0002]

[Description of the Prior Art] Heat-sealing nature of an ionomer is good, therefore it is advantageously used as a material for a technical field, for example, a meat package, with which heat-sealing nature is demanded. However, in view of the various demand characteristics in an application technical field, if an ionomer is usually independent, it is not used, but it becomes together with other materials, for example, is used in the state of a layered product. What pasted up the ionomer layer and the ethylene-vinylalcohol copolymer layer as a layered product, and pasted up the polyethylene system adhesive property resin layer as an interlayer, for example is known. However, interlaminar peeling occurred or there was a fault which an adhesive property with an ionomer has, and is not enough as the thermal resistance at the time of heat-treatment. [ of conventional polyethylene system adhesive property resin ] [ inadequate ]

[0003]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the layered product which has the resin constituent layer excellent in an adhesive property and thermal resistance with an ionomer. Other purposes of this invention are to offer the layered product which has the heat-sealing nature which did not produce interlaminar peeling and was excellent. The purpose of further others of this invention is to offer the layered product which has oxygen barrier nature in addition to the outstanding property like the above. The purpose and advantage of further others of this invention will become clear from the following explanation.

[0004]

[Means for Solving the Problem] According to this invention, the above-mentioned purpose and advantage of this invention The ionomer layer set to the 1st from (1) and (1) ionomer, In a list, according to the (2) and (A) transition metal catalyst The crystalline ethylene and alpha olefin copolymer, and the (b) consistency whose (a) consistency by which the polymerization was carried out is 0.89 - 0.94 g/cm<sup>3</sup> are less than [ 0.89g //cm ] three, and crystallinity consists of 40% or less of amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity. The crystalline ethylene and the alpha olefin copolymer the 1st resin constituent whose consistencies are less than three 0.930 g/cm<sup>3</sup>, and whose (c) consistency the polymerization was carried out [ the consistency ] by the (B) transition metal catalyst are 0.92 g/cm<sup>3</sup> at least, (d) Ethylene-vinylacetate copolymer, From the ethylene copolymer chosen from the group which consists of an ethylene acrylic-acid copolymer and an ethylene ethyl acrylate copolymer The 2nd resin constituent which contains (c) crystallinity ethylene and 95 - 50 % of the weight of alpha olefin copolymers, and 5 - 50 % of the weight of (d) ethylene copolymers based on the sum total weight of crystalline ethylene and the alpha olefin copolymer (above-mentioned

[ c ]), and the ethylene copolymer (above-mentioned [ d ]) And it is attained by the resin layered product characterized by the resin constituent layer which consists of a resin constituent chosen from the group which consists of the 3rd resin constituent which comes to carry out graft denaturation of the 1st resin constituent of (C) above or the above-mentioned 2nd resin constituent with unsaturated carboxylic acid adjoining and existing.

[0005] The resin layered product of this invention is characterized by the ionomer layer which consists of (1) ionomer, and the resin constituent layer chosen from the (2) 1st, 2nd, and 3rd resin constituent adjoining and existing as above-mentioned.

[0006] As an ionomer which forms an ionomer layer (1), that in which a part of carboxyl group of a copolymer with alpha like ethylene, an acrylic acid, or a methacrylic acid and beta-unsaturated carboxylic acid forms the salt with alkali metal or alkaline earth metal, for example is desirable. The copolymerization percentage of alpha and beta-unsaturated carboxylic acid is 0.1-30-mol % preferably. Na salt, a calcium salt, magnesium salt, or Zn salt is used advantageously.

[0007] The crystalline ethylene and alpha olefin copolymer, and the (b) consistency whose (a) consistency to which the polymerization of the 1st resin constituent which forms a resin constituent layer (2) was carried out by the transition metal catalyst is  $0.89 - 0.94 \text{ g/cm}^3$  are less than  $[ 0.89 \text{ g/cm}^3 ]$  three, and crystallinity consists of 40% or less of amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity. Here, a tsi GURA type, Philip SUTAIPU, and a metallocene type are illustrated as the transition metal catalyst used by the polymerization.

[0008] (a) Crystalline ethylene and an alpha olefin copolymer are random copolymers of ethylene and the alpha olefin of carbon numbers 3-20 preferably. As an alpha olefin, the alpha olefin of carbon numbers 4-10 is [ among these ] desirable. As an alpha olefin, a propylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene, etc. can be mentioned as a desirable thing. Moreover, (a) ethylene and an alpha olefin copolymer contain more preferably the polymerization unit which originates in ethylene preferably at 99 - 85 % of the weight 99 to 80% of the weight. The consistency of ethylene and an alpha olefin copolymer is three or less  $[ 3 \text{ or more } \text{g/cm}^3 / 0.940 ] 0.89 \text{ g/cm}^3$ , and is  $[ \text{cm}^3 ] 3 0.900 - 0.935 \text{ g/cm}^3$  preferably. Moreover, the degree of crystallinity (it measures with an X-ray diffraction method) of ethylene and an alpha olefin copolymer is larger than 40%, and is 55 - 80% preferably. This (a) ethylene and alpha olefin copolymer have MI value in the range for 0.1 - 30g / 10 minutes (190 degrees C) preferably.

[0009] Another (b) amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity is already a random copolymer of beam ethylene and the alpha olefin of carbon numbers 3-20 preferably. The thing same as an alpha olefin as what was mentioned above about said ethylene and alpha olefin copolymer can be mentioned. The alpha olefin of carbon numbers 3 or 4 is [ among these ] desirable. (b) Ethylene and an alpha olefin copolymer contain the polymerization unit which originates in ethylene preferably at 75-90-mol % more preferably than 75-95-mol %. (b) The consistency of ethylene and an alpha olefin copolymer is less than three  $0.89 \text{ g/cm}^3$ , and is less than three three or more  $0.85 \text{ g/cm}^3 0.89 \text{ g/cm}^3$  preferably. Moreover, the degree of crystallinity of (b) ethylene and an alpha olefin copolymer is 40% or less, and is 30% or less preferably. This (b) ethylene and alpha olefin copolymer have MI value in the range for 0.1 - 50g / 10 minutes (190 degrees C) preferably.

[0010] In the 1st resin constituent, (a) crystallinity ethylene and an alpha olefin copolymer, (b) amorphism nature, or low crystallinity ethylene and an alpha olefin copolymer has the desirable rate that (a) crystallinity ethylene and an alpha olefin copolymer occupy 99 - 50 % of the weight based on these sum total polymerizations, and (b) amorphism nature, or low crystallinity ethylene and an alpha olefin copolymer occupies 5 - 45 % of the weight more preferably.

[0011] The consistencies of the 1st resin constituent are less than three  $0.930 \text{ g/cm}^3$ , and are three or less  $[ 0.920 \text{ g/cm}^3 ]$  preferably. Moreover, the 2nd resin constituent which forms a resin constituent layer (2) consists of the crystalline ethylene and alpha olefin copolymer, and (d) ethylene copolymer whose (c) consistencies the polymerization was carried out [ the consistencies ] by the transition metal catalyst are  $0.920 \text{ g/cm}^3$  at least. The consistency of the crystalline ethylene and the alpha olefin copolymer of  $0.920 \text{ g/cm}^3$  (c) is the random copolymer of ethylene and the alpha olefin of carbon numbers 3-20 preferably

at least. The same thing as what was mentioned above about said ethylene and alpha olefin copolymer as an alpha olefin can be mentioned. Ethylene and an alpha olefin copolymer (c) contain the polymerization unit originating in ethylene at 99 - 85 % of the weight preferably more preferably than 99 - 80 % of the weight. The consistencies of ethylene and an alpha olefin copolymer (c) are 0.920 g/cm<sup>3</sup> at least, and are 0.920 - 0.935 g/cm<sup>3</sup> preferably. (d) An ethylene copolymer is chosen from the group which consists of an ethylene-vinyl acetate copolymer, an ethylene acrylic-acid copolymer, and an ethylene ethyl acrylate copolymer. An ethylene-vinylacetate copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. An ethylene acrylic-acid copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. An ethylene ethyl acrylate copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. the 2nd resin constituent -- the (c) consistency -- at least -- the sum total weight of the crystalline ethylene and the alpha olefin copolymer of 0.92g/cm<sup>3</sup>, and (d) ethylene copolymer -- being based -- crystalline ethylene and the alpha olefin copolymer (above-mentioned [ c ]) -- 95 - 50 % of the weight -- it contains ten to 40% of the weight preferably 5 - 50 % of the weight of 90 - 60 % of the weight (d) ethylene copolymers. Furthermore, the 3rd resin constituent which forms a resin constituent layer (2) comes to carry out graft denaturation of the above-mentioned 1st resin constituent or the 2nd resin constituent with unsaturated carboxylic acid. As partial saturation carvones, unsaturated carboxylic acid or its derivative is used.

[0012] As unsaturated carboxylic acid or its derivative, unsaturated-carboxylic-acid [, such as an acrylic acid, a maleic acid, boletic acid, a tetrahydrophthal acid, an itaconic acid, a citraconic acid, a crotonic acid isocrotonic acid, and a NAJIKKU acid (the endo-cis-bicyclo [2.2.1] hept-5-en -2, 3-dicarboxylic acid), ]; or its derivative, for example, acid halide, an amide, imide, an anhydride, ester, etc. are mentioned, for example. As an example of this derivative, chlorination MARENIRU, maleimide, a maleic anhydride, an anhydrous citraconic acid, maleic-acid monomethyl, maleic-acid dimethyl, glycidil maleate, etc. are mentioned, for example. In these, partial saturation dicarboxylic acid or its acid anhydride is suitable, and a maleic acid, NAJIKKU acids, or these acid anhydrides are used especially preferably.

[0013] To the 1st resin constituent or the 2nd resin constituent, the graft denaturation of the 1st resin constituent and the 2nd resin constituent is good in a direct line, or may be performed to the one section or all of each component that constitutes the 1st resin constituent and the 2nd resin constituent. As for graft denaturation, it is desirable to carry out about the ethylene and the alpha olefin copolymer which constitutes each resin constituent. Based on the ethylene and the alpha olefin copolymer before denaturation, graft denaturation of the ethylene and the alpha olefin copolymer which denaturalized with unsaturated carboxylic acid or its derivative is more preferably carried out 0.001 to 15% of the weight with 0.01 - 10% of the weight of unsaturated carboxylic acid, or its derivative.

[0014] The laminating structure of this invention can have the layer of further others besides the above-mentioned ionomer layer and a resin constituent layer. Other layers can be polyethylene, an ethylene-vinylalcohol copolymer, polyester, or a polyamide (nylon). As for other layers, it is desirable to adjoin a resin constituent layer. When other layers are ethylene vinyl alcohol, it is suitable for a resin constituent layer to consist of the above-mentioned 3rd resin constituent.

[0015] The resin layer chosen from the group which consists of the ionomer layer which consists of (1) ionomer, the resin constituent layer which consists of the (2) 3rd resin constituent, (3) nylon, and an ethylene-vinylalcohol copolymer as one desirable mode adjoins in such sequence, and the resin structure of this invention exists.

[0016] The layered product of this invention can fuse the resin or the resin constituent which constitutes each class, respectively, and can manufacture it by carrying out a laminating in the state of melting. The laminating structure can be fabricated in a desired gestalt by extrusion molding, cast shaping, inflation molding, etc.

[0017]

[Example] This invention is further explained in full detail according to an example below. However,

this invention is not limited to these.

[0018] The ethylene and the 4-methyl-1-pentene copolymer of consistency 0.920 g/cm<sup>3</sup> by which the polymerization was carried out with the example 1 tsi GURA type catalyst (MI=2.0g /, and ten parts (190 degrees C)) 90 weight sections called LL-1 for short below and ethylene propylene rubber of consistency 0.88 g/cm<sup>3</sup> (MI=1.1g /, and ten parts) Melting mixing of ethylene content % and 0% of degree of crystallinity of 80 mols, and the 10 weight sections called EPR-1 for short below was carried out at 210 degrees C with 1 shaft extruder, and the adhesive resin constituent was obtained. The consistencies of this adhesive constituent were 0.916 g/cm<sup>3</sup>, and MI values were 1.80g / 10 minutes (190 degrees C). An ionomer (yes, milan 1601, Na type, Product made from Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and low density polyethylene (made in [ Mitsui Petrochemical Industries, Ltd. ] Myra Son F967) are prepared. Each is fused at 200 degrees C, 210 degrees C, and 210 degrees C by this sequence. By 20m a part for /and the blow up ratio 1.0 in shaping rate An ionomer layer / adhesive resin constituent layer / low-density-polyethylene layer = the three-layer laminated film (40 micrometers / 10 micrometers / 40 micrometers) was manufactured by the air-cooling tubular film process. The adhesive strength between the ionomer layer of this laminated film and an adhesive resin constituent layer was 790g / 15mm.

[0019] In examples 2-6 and one to example of comparison 2 example 1, it changed into the presentation which showed the presentation of an adhesive resin constituent in Table 1, and also the three-layer laminating air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 1. In addition, the data about an example 1 were also shown in Table 1.

[0020]

[Table 1]

表 1

実施例	1	2	3	4	5	6	比較例 1	比較例 2
組成と重量部	LL-1 90 EPR-1 10	LL-1 80 EPR-1 20	LL-1 70 EPR-1 30	LL-1 90 EPR-1 10	LL-1 80 EPR-1 20	LL-1 70 EPR-1 30	LL-1 100	PE-1 80 EPR-1 20
接着性樹脂組成物	MI 190°C (g/10min)	1.80	1.70	1.55	2.10	2.25	2.40	1.90
密度 (g/cm <sup>3</sup> )	0.916	0.911	0.906	0.919	0.915	0.912	0.920	0.912
接着力 (g/15mm)	790	800	870	740	800	830	210	460

注) LL-1 : エチレン-4-メチル-1-ペンテン共重合体

(MI=2.0g/10分、密度0.920g/cm<sup>3</sup>)

EPR-1 : エチレン・プロピレン共重合体

(MI=1.0g/10分、密度0.88g/cm<sup>3</sup>、エチレン含量80wt%)

EBR-1 : エチレン-1-ブテン共重合体

(MI=3.6g/10分、密度0.885g/cm<sup>3</sup>、エチレン含量90wt%)

PE-1 : 高圧法低密度ポリエチレン

(MI=3.6g/10分、密度0.923g/cm<sup>3</sup>)

[0021] It changed into the presentation which showed the presentation of the adhesive resin constituent in seven to example 11 example 1 in Table 2, and also the three-layer air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 2.

[0022]

[Table 2]

表 2

実施例	7	8	9	10	11
接着性樹脂組成物	LL-1 90 EVA-1 10	LL-1 80 EVA-1 20	LL-1 70 EVA-1 30	LL-1 80 EPR-1 10 EVA-1 10	LL-1 80 EEA-1 20
MI 190℃ (g/10min)	2.00	2.00	2.00	1.85	2.20
密度 (g/cm <sup>3</sup> )	0.924	0.927	0.929	0.919	0.922
接着力 (g/15mm)	770	760	720	750	780

注) LL-1 : エチレン・4-メチル-1-ペンテン共重合体

(MI=2.0g/10分、密度=0.920g/cm<sup>3</sup>)

EVA-1 : エチレン・酢酸ビニル共重合体

(MI=2g/10分、密度=0.95g/cm<sup>3</sup>、酢酸ビニル含量25重量%)

EEA-1 : エチレン・エチルアクリレート共重合体

(MI=5g/10分、エチルアクリレート含量25重量%)

EPR-1 : エチレン・プロピレン共重合体

(MI=1.0g/10分、密度=0.88g/cm<sup>3</sup>、エチレン含量80重量%)

[0023] The ethylene and 1-butene copolymer of consistency 0.923 g/cm<sup>3</sup> by which the polymerization was carried out with the example 12 tsi GURA type catalyst (MI=2.0g /, and ten parts (190 degrees C)) 82 weight sections called LL-2 for short below and ethylene propylene rubber of consistency 0.88 g/cm<sup>3</sup> (MI=2.9g /, and ten parts) Ethylene content % and 0% of degree of crystallinity of 80 mols, 15 weight sections called EPR-2 for short below, And melting mixing of the maleic-acid graft denaturation polyolefine for MI=4g /, and 10 minutes (the rate of a maleic-acid graft = it calls for short Following MAHPE 2% of the weight) was carried out at 210 degrees C with 1 shaft extruder, and the adhesive resin constituent was obtained. The consistencies of this adhesive resin constituent were 0.916 g/cm<sup>3</sup>. An ionomer (yes, Product made from milan 1601 (Na type) Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and nylon (Amilan CM1021XF Toray Industries, Inc. make) were fused at 200 degrees C, 210 degrees C, and 260 degrees C by this sequence, respectively, and the three-layer laminated film (an ionomer layer / adhesive resin constituent layer / nylon layer =40micrometer / 10 micrometers / 20 micrometers) was obtained by the air-cooling tubular film process by shaping rate 20 m/min and the blow up ratio 1.0. The adhesive strength of the interface of the ionomer layer of this laminated film and an adhesive resin constituent layer and the adhesive strength of the interface of nylon and an adhesive resin constituent layer cannot be exfoliated [ 700g / 15mm, and ], respectively. Moreover, the ionomer (yes, Product made from milan 1605 (Zn type) Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and the ethylene-vinylalcohol copolymer (Eval EP-F101A, Kuraray Co., Ltd. make) were fused at 200 degrees C, 210 degrees C, and 210 degrees C by this sequence, respectively, and the three-layer laminated film (an ionomer layer / adhesive resin constituent layer / ethylene-vinylalcohol copolymer layer =40micrometer / 10 micrometers / 20 micrometers) was obtained by the air-cooling tubular film process by shaping rate 20 m/min and the blow up ratio 1.0. The adhesive strength of the interface of the ionomer layer of this laminated film and an adhesive resin constituent layer and the adhesive strength of the interface of an ethylene-vinylalcohol copolymer layer and an adhesive resin constituent layer were 770g / 15mm, and 500g / 15mm, respectively.

[0024] It changed into the presentation which showed the presentation of the adhesive resin constituent in examples 13-15 and three to example of comparison 4 example 12 in Table 3, and also the three-layer air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 3.

[0025]

[Table 3]

表 3

実 施 例		1 2	1 3	1 4	1 5	比較例 3	比較例 4
接着性樹脂組成物	組成と重量部	LL-2 82 EPR-2 15 MAHPE 3	LL-2 80 EPR-2 15 MAHPE 5	LL-2 65 EPR-2 30 MAHPE 5	LL-2 52 EPR-2 45 MAHPE 3	HDPE 98 MAHPE 2	LDPE 80 EPR-1 15 MAHPE 5
	M I 190℃ (g/10min)	2.0	2.0	2.1	2.2	1.0	1.5
	密度 (g/cm <sup>3</sup> )	0.916	0.916	0.908	0.900	0.966	0.912
	接着力 (g/15mm)						
	ハイミラン 1601	700	720	710	710	40	320
	タイロン	剝離不能	剝離不能	剝離不能	剝離不能	800	840
	ハイミラン 1605	770	780	770	760	50	500
	EVOH	500	650	780	680	20	700

注) LL-2 : エチレン・1-ブテン共重合体 (MI=2.0g/10分、密度=0.923g/cm<sup>3</sup>)

EPR-2 : エチレン・プロピレン共重合体  
(MI=2.9g/10分、密度=0.88g/cm<sup>3</sup>、エチレン含量80%)

MAHPE : マレイン酸グラフト変性ポリエチレン  
(MI=4g/10分、マレイン酸グラフト率2重量%)

HDPE-1 : 高密度ポリエチレン (MI=1g/10分、密度0.966g/15mm)

PE-2 : 高圧法低密度ポリエチレン (MI=2g/10分、密度0.920g/cm<sup>3</sup>)

タイロン : 東レ(株)製 アミロンCM1021XF

EVOH : クラレ(株)製 エボールEP-F101A

[0026]

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**PRIOR ART**

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[Description of the Prior Art] Heat-sealing nature of an ionomer is good, therefore it is advantageously used as a material for a technical field, for example, a meat package, with which heat-sealing nature is demanded. However, in view of the various demand characteristics in an application technical field, if an ionomer is usually independent, it is not used, but it becomes together with other materials, for example, is used in the state of a layered product. What pasted up the ionomer layer and the ethylene-vinylalcohol copolymer layer as a layered product, and pasted up the polyethylene system adhesive property resin layer as an interlayer, for example is known. However, interlaminar peeling occurred or there was a fault which an adhesive property with an ionomer has, and is not enough as the thermal resistance at the time of heat-treatment. [ of conventional polyethylene system adhesive property resin ] [ inadequate ]

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**TECHNICAL PROBLEM**

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[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the layered product which has the resin constituent layer excellent in an adhesive property and thermal resistance with an ionomer. Other purposes of this invention are to offer the layered product which has the heat-sealing nature which did not produce interlaminar peeling and was excellent. The purpose of further others of this invention is to offer the layered product which has oxygen barrier nature in addition to the outstanding property like the above. The purpose and advantage of further others of this invention will become clear from the following explanation.

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**MEANS**

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[Means for Solving the Problem] According to this invention, the above-mentioned purpose and advantage of this invention The ionomer layer set to the 1st from (1) and (1) ionomer, In a list, according to the (2) and (A) transition metal catalyst The crystalline ethylene and alpha olefin copolymer, and the (b) consistency whose (a) consistency by which the polymerization was carried out is 0.89 - 0.94 g/cm<sup>3</sup> are less than [ 0.89g //cm ] three, and crystallinity consists of 40% or less of amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity. The crystalline ethylene and the alpha olefin copolymer the 1st resin constituent whose consistencies are less than three 0.930 g/cm<sup>3</sup>, and whose (c) consistency the polymerization was carried out [ the consistency ] by the (B) transition metal catalyst are 0.92 g/cm<sup>3</sup> at least, (d) Ethylene-vinylacetate copolymer, From the ethylene copolymer chosen from the group which consists of an ethylene acrylic-acid copolymer and an ethylene ethyl acrylate copolymer The 2nd resin constituent which contains (c) crystallinity ethylene and 95 - 50 % of the weight of alpha olefin copolymers, and 5 - 50 % of the weight of (d) ethylene copolymers based on the sum total weight of crystalline ethylene and the alpha olefin copolymer (above-mentioned [ c ]), and the ethylene copolymer (above-mentioned [ d ]) And it is attained by the resin layered product characterized by the resin constituent layer which consists of a resin constituent chosen from the group which consists of the 3rd resin constituent which comes to carry out graft denaturation of the 1st resin constituent of (C) above or the above-mentioned 2nd resin constituent with unsaturated carboxylic acid adjoining and existing.

[0005] The resin layered product of this invention is characterized by the ionomer layer which consists of (1) ionomer, and the resin constituent layer chosen from the (2) 1st, 2nd, and 3rd resin constituent adjoining and existing as above-mentioned.

[0006] As an ionomer which forms an ionomer layer (1), that in which a part of carboxyl group of a copolymer with alpha like ethylene, an acrylic acid, or a methacrylic acid and beta-unsaturated carboxylic acid forms the salt with alkali metal or alkaline earth metal, for example is desirable. The copolymerization percentage of alpha and beta-unsaturated carboxylic acid is 0.1-30-mol % preferably. Na salt, a calcium salt, magnesium salt, or Zn salt is used advantageously.

[0007] The crystalline ethylene and alpha olefin copolymer, and the (b) consistency whose (a) consistency to which the polymerization of the 1st resin constituent which forms a resin constituent layer (2) was carried out by the transition metal catalyst is 0.89 - 0.94 g/cm<sup>3</sup> are less than [ 0.89g //cm ] three, and crystallinity consists of 40% or less of amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity. Here, a tsi GURA type, Philip SUTAIPU, and a metallocene type are illustrated as the transition metal catalyst used by the polymerization.

[0008] (a) Crystalline ethylene and an alpha olefin copolymer are random copolymers of ethylene and the alpha olefin of carbon numbers 3-20 preferably. As an alpha olefin, the alpha olefin of carbon numbers 4-10 is [ among these ] desirable. As an alpha olefin, a propylene, 1-butene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene, etc. can be mentioned as a desirable thing. Moreover, (a) ethylene and an alpha olefin copolymer contain more preferably the polymerization unit which originates in ethylene preferably at 99 - 85 % of the weight 99 to 80% of the weight. The consistency of ethylene and

an alpha olefin copolymer is three or less [ 3 or more g //cm / 0.940 ] 0.89 g/cm, and is [ cm ] 3 0.900-0.935g /preferably. Moreover, the degree of crystallinity (it measures with an X-ray diffraction method) of ethylene and an alpha olefin copolymer is larger than 40%, and is 55 - 80% preferably. This (a) ethylene and alpha olefin copolymer have MI value in the range for 0.1 - 30g / 10 minutes (190 degrees C) preferably.

[0009] Another (b) amorphism nature, or the ethylene and the alpha olefin copolymer of low crystallinity is already a random copolymer of beam ethylene and the alpha olefin of carbon numbers 3-20 preferably. The thing same as an alpha olefin as what was mentioned above about said ethylene and alpha olefin copolymer can be mentioned. The alpha olefin of carbon numbers 3 or 4 is [ among these ] desirable. (b) Ethylene and an alpha olefin copolymer contain the polymerization unit which originates in ethylene preferably at 75-90-mol % more preferably than 75-95-mol %. (b) The consistency of ethylene and an alpha olefin copolymer is less than three 0.89 g/cm, and is less than three three or more 0.85 g/cm0.89 g/cm preferably. Moreover, the degree of crystallinity of (b) ethylene and an alpha olefin copolymer is 40% or less, and is 30% or less preferably. This (b) ethylene and alpha olefin copolymer have MI value in the range for 0.1 - 50g / 10 minutes (190 degrees C) preferably.

[0010] In the 1st resin constituent, (a) crystallinity ethylene and an alpha olefin copolymer, (b) amorphism nature, or low crystallinity ethylene and an alpha olefin copolymer has the desirable rate that (a) crystallinity ethylene and an alpha olefin copolymer occupy 99 - 50 % of the weight based on these sum total polymerizations, and (b) amorphism nature, or low crystallinity ethylene and an alpha olefin copolymer occupies 5 - 45 % of the weight more preferably.

[0011] The consistencies of the 1st resin constituent are less than three 0.930 g/cm, and are three or less [ 0.920g //cm ] preferably. Moreover, the 2nd resin constituent which forms a resin constituent layer (2) consists of the crystalline ethylene and alpha olefin copolymer, and (d) ethylene copolymer whose (c) consistencies the polymerization was carried out [ the consistencies ] by the transition metal catalyst are 0.920 g/cm<sup>3</sup> at least. The consistency of the crystalline ethylene and the alpha olefin copolymer of 0.920 g/cm<sup>3</sup> (c) is the random copolymer of ethylene and the alpha olefin of carbon numbers 3-20 preferably at least. The same thing as what was mentioned above about said ethylene and alpha olefin copolymer as an alpha olefin can be mentioned. Ethylene and an alpha olefin copolymer (c) contain the polymerization unit originating in ethylene at 99 - 85 % of the weight preferably more preferably than 99 - 80 % of the weight. The consistencies of ethylene and an alpha olefin copolymer (c) are 0.920 g/cm<sup>3</sup> at least, and are 0.920 - 0.935 g/cm<sup>3</sup> preferably. (d) An ethylene copolymer is chosen from the group which consists of an ethylene-vinyl acetate copolymer, an ethylene acrylic-acid copolymer, and an ethylene ethyl acrylate copolymer. An ethylene-vinylacetate copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. An ethylene acrylic-acid copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. An ethylene ethyl acrylate copolymer contains more preferably the polymerization unit which originates in ethylene preferably 70 to 95% of the weight 65 to 95% of the weight. the 2nd resin constituent -- the (c) consistency -- at least -- the sum total weight of the crystalline ethylene and the alpha olefin copolymer of 0.92g/cm<sup>3</sup>, and (d) ethylene copolymer -- being based -- crystalline ethylene and the alpha olefin copolymer (above-mentioned [ c ]) -- 95 - 50 % of the weight -- it contains ten to 40% of the weight preferably 5 - 50 % of the weight of 90 - 60 % of the weight (d) ethylene copolymers. Furthermore, the 3rd resin constituent which forms a resin constituent layer (2) comes to carry out graft denaturation of the above-mentioned 1st resin constituent or the 2nd resin constituent with unsaturated carboxylic acid. As partial saturation carvones, unsaturated carboxylic acid or its derivative is used.

[0012] As unsaturated carboxylic acid or its derivative, unsaturated-carboxylic-acid [ , such as an acrylic acid, a maleic acid, boletic acid, a tetrahydrophtal acid, an itaconic acid, a citraconic acid, a crotonic acid isocrotonic acid, and a NAJIKKU acid (the endo-cis-bicyclo [2.2.1] hept-5-en -2, 3-dicarboxylic acid), ]; or its derivative, for example, acid halide, an amide, imide, an anhydride, ester, etc. are mentioned, for example. As an example of this derivative, chlorination MARENIRU, maleimide, a maleic anhydride, an anhydrous citraconic acid, maleic-acid monomethyl, maleic-acid dimethyl, glycidil

maleate, etc. are mentioned, for example. In these, partial saturation dicarboxylic acid or its acid anhydride is suitable, and a maleic acid, NAJIKKU acids, or these acid anhydrides are used especially preferably.

[0013] To the 1st resin constituent or the 2nd resin constituent, the graft denaturation of the 1st resin constituent and the 2nd resin constituent is good in a direct line, or may be performed to the one section or all of each component that constitutes the 1st resin constituent and the 2nd resin constituent. As for graft denaturation, it is desirable to carry out about the ethylene and the alpha olefin copolymer which constitutes each resin constituent. Based on the ethylene and the alpha olefin copolymer before denaturation, graft denaturation of the ethylene and the alpha olefin copolymer which denaturalized with unsaturated carboxylic acid or its derivative is more preferably carried out 0.001 to 15% of the weight with 0.01 - 10% of the weight of unsaturated carboxylic acid, or its derivative.

[0014] The laminating structure of this invention can have the layer of further others besides the above-mentioned ionomer layer and a resin constituent layer. Other layers can be polyethylene, an ethylene-vinylalcohol copolymer, polyester, or a polyamide (nylon). As for other layers, it is desirable to adjoin a resin constituent layer. When other layers are ethylene vinyl alcohol, it is suitable for a resin constituent layer to consist of the above-mentioned 3rd resin constituent.

[0015] The resin layer chosen from the group which consists of the ionomer layer which consists of (1) ionomer, the resin constituent layer which consists of the (2) 3rd resin constituent, (3) nylon, and an ethylene-vinylalcohol copolymer as one desirable mode adjoins in such sequence, and the resin structure of this invention exists.

[0016] The layered product of this invention can fuse the resin or the resin constituent which constitutes each class, respectively, and can manufacture it by carrying out a laminating in the state of melting. The laminating structure can be fabricated in a desired gestalt by extrusion molding, cast shaping, inflation molding, etc.

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[Translation done.]

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EXAMPLE

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[Example] This invention is further explained in full detail according to an example below. However, this invention is not limited to these.

[0018] The ethylene and the 4-methyl-1-pentene copolymer of consistency 0.920 g/cm<sup>3</sup> by which the polymerization was carried out with the example 1 tsi GURA type catalyst (MI=2.0g /, and ten parts (190 degrees C)) 90 weight sections called LL-1 for short below and ethylene propylene rubber of consistency 0.88 g/cm<sup>3</sup> (MI=1.1g /, and ten parts) Melting mixing of ethylene content % and 0% of degree of crystallinity of 80 mols, and the 10 weight sections called EPR-1 for short below was carried out at 210 degrees C with 1 shaft extruder, and the adhesive resin constituent was obtained. The consistencies of this adhesive constituent were 0.916 g/cm<sup>3</sup>, and MI values were 1.80g / 10 minutes (190 degrees C). An ionomer (yes, milan 1601, Na type, Product made from Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and low density polyethylene (made in [ Mitsui Petrochemical Industries, Ltd. ] Myra Son F967) are prepared. Each is fused at 200 degrees C, 210 degrees C, and 210 degrees C by this sequence. By 20m a part for /and the blow up ratio 1.0 in shaping rate An ionomer layer / adhesive resin constituent layer / low-density-polyethylene layer = the three-layer laminated film (40 micrometers / 10 micrometers / 40 micrometers) was manufactured by the air-cooling tubular film process. The adhesive strength between the ionomer layer of this laminated film and an adhesive resin constituent layer was 790g / 15mm.

[0019] In examples 2-6 and one to example of comparison 2 example 1, it changed into the presentation which showed the presentation of an adhesive resin constituent in Table 1, and also the three-layer laminating air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 1. In addition, the data about an example 1 were also shown in Table 1.

[0020]

[Table 1]

表 1

実施例	1	2	3	4	5	6	比較例 1	比較例 2
組成と重量部	LL-1 90 EPR-1 10	LL-1 80 EPR-1 20	LL-1 70 EPR-1 30	LL-1 90 EPR-1 10	LL-1 80 EPR-1 20	LL-1 70 EPR-1 30	LL-1 100	PE-1 80 EPR-1 20
接着性	1.80	1.70	1.55	2.10	2.25	2.40	2.00	1.90
樹脂組成物	MI 190°C (g/10min)							
密度 (g/cm <sup>3</sup> )	0.916	0.911	0.906	0.919	0.915	0.912	0.920	0.912
接着力 (g/15mm)	790	800	870	740	800	830	210	460

注) LL-1 : エチレン-4-メチル-1-ペンテン共重合体  
(MI=2.0g/10分、密度0.920g/cm<sup>3</sup>)

EPR-1 : エチレン・プロピレン共重合体  
(MI=1.0g/10分、密度0.88g/cm<sup>3</sup>、エチレン含量80wt%)

EBR-1 : エチレン-1-ブテン共重合体  
(MI=3.6g/10分、密度0.885g/cm<sup>3</sup>、エチレン含量90wt%)

PE-1 : 高圧法低密度ポリエチレン  
(MI=3.6g/10分、密度0.923g/cm<sup>3</sup>)

[0021] It changed into the presentation which showed the presentation of the adhesive resin constituent in seven to example 11 example 1 in Table 2, and also the three-layer air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 2.

[0022]

[Table 2]

表 2

実施例	7	8	9	10	11
接着性樹脂組成物	LL-1 90 EVA-1 10	LL-1 80 EVA-1 20	LL-1 70 EVA-1 30	LL-1 80 EPR-1 10 EVA-1 10	LL-1 80 EEA-1 20
MI 190℃ (g/10min)	2.00	2.00	2.00	1.85	2.20
密度 (g/cm <sup>3</sup> )	0.924	0.927	0.929	0.919	0.922
接着力 (g/15mm)	770	760	720	750	780

注) LL-1 : エチレン・4-メチル-1-ペンテン共重合体

(MI=2.0g/10分、密度=0.920g/cm<sup>3</sup>)

EVA-1 : エチレン・酢酸ビニル共重合体

(MI=2g/10分、密度=0.95g/cm<sup>3</sup>、酢酸ビニル含量25重量%)

EEA-1 : エチレン・エチルアクリレート共重合体

(MI=5g/10分、エチルアクリレート含量25重量%)

EPR-1 : エチレン・プロピレン共重合体

(MI=1.0g/10分、密度=0.88g/cm<sup>3</sup>、エチレン含量80重量%)

[0023] The ethylene and 1-butene copolymer of consistency 0.923 g/cm<sup>3</sup> by which the polymerization was carried out with the example 12 tsi GURA type catalyst (MI=2.0g /, and ten parts (190 degrees C)) 82 weight sections called LL-2 for short below and ethylene propylene rubber of consistency 0.88 g/cm<sup>3</sup> (MI=2.9g /, and ten parts) Ethylene content % and 0% of degree of crystallinity of 80 mols, 15 weight sections called EPR-2 for short below, And melting mixing of the maleic-acid graft denaturation polyolefine for MI=4g /, and 10 minutes (the rate of a maleic-acid graft = it calls for short Following MAHPE 2% of the weight) was carried out at 210 degrees C with 1 shaft extruder, and the adhesive resin constituent was obtained. The consistencies of this adhesive resin constituent were 0.916 g/cm<sup>3</sup>. An ionomer (yes, Product made from milan 1601 (Na type) Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and nylon (Amilan CM1021XF Toray Industries, Inc. make) were fused at 200 degrees C, 210 degrees C, and 260 degrees C by this sequence, respectively, and the three-layer laminated film (an ionomer layer / adhesive resin constituent layer / nylon layer =40micrometer / 10 micrometers / 20 micrometers) was obtained by the air-cooling tubular film process by shaping rate 20 m/min and the blow up ratio 1.0. The adhesive strength of the interface of the ionomer layer of this laminated film and an adhesive resin constituent layer and the adhesive strength of the interface of nylon and an adhesive resin constituent layer cannot be exfoliated [ 700g / 15mm, and ], respectively. Moreover, the ionomer (yes, Product made from milan 1605 (Zn type) Mitsui DEYUPON poly chemical), the above-mentioned adhesive resin constituent, and the ethylene-vinylalcohol copolymer (Eval EP-F101A, Kuraray Co., Ltd. make) were fused at 200 degrees C, 210 degrees C, and 210 degrees C by this sequence, respectively, and the three-layer laminated film (an ionomer layer / adhesive resin constituent layer / ethylene-vinylalcohol copolymer layer =40micrometer / 10 micrometers / 20 micrometers) was obtained by the air-cooling tubular film process by shaping rate 20 m/min and the blow up ratio 1.0. The adhesive strength of the interface of the ionomer layer of this laminated film and an adhesive resin constituent layer and the adhesive strength of the interface of an ethylene-vinylalcohol copolymer layer and an adhesive resin constituent layer were 770g / 15mm, and 500g / 15mm, respectively.

[0024] It changed into the presentation which showed the presentation of the adhesive resin constituent in examples 13-15 and three to example of comparison 4 example 12 in Table 3, and also the three-layer air-cooling tubular blown film was obtained like the example 1. A result is shown in Table 3.

[0025]

[Table 3]



表 3

実施例		1 2	1 3	1 4	1 5	比較例 3	比較例 4
接着性樹脂組成物	組成と重量部	LL-2 82 EPR-2 15 MAHPE 3	LL-2 80 EPR-2 15 MAHPE 5	LL-2 65 EPR-2 30 MAHPE 5	LL-2 52 EPR-2 45 MAHPE 3	HDPE 98 MAHPE 2	LDPE 80 EPR-1 15 MAHPE 5
	MI 190℃ (g/10min)	2.0	2.0	2.1	2.2	1.0	1.5
	密度 (g/cm <sup>3</sup> )	0.916	0.916	0.908	0.900	0.966	0.912
接着力 (g/15mm)	ハイミラン 1601	700	720	710	710	40	320
	タイロン	剥離不能	剥離不能	剥離不能	剥離不能	800	840
	ハイミラン 1605	770	780	770	760	50	500
	EVOH	500	650	780	680	20	700

注) LL-2 : エチレン・1-ブテン共重合体 (MI=2.0g/10分、密度=0.923g/cm<sup>3</sup>)  
 EPR-2 : エチレン・プロピレン共重合体  
 (MI=2.9g/10分、密度=0.88g/cm<sup>3</sup>、エチレン含量80wt%)  
 MAHPE : マレイン酸グラフト変性ポリエチレン  
 (MI=4g/10分、マレイン酸グラフト率2重量%)  
 HDPE-1 : 高密度ポリエチレン (MI=1g/10分、密度0.966g/15mm)  
 PE-2 : 高圧法低密度ポリエチレン (MI=2g/10分、密度0.920g/cm<sup>3</sup>)  
 タイロン : 東レ(株)製 タイロンCM1021XF  
 EVOH : クラレ(株)製 EVOH-EP-F101A

[Translation done.]